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Investigations of ultra short femtosecond pulse propagation of > 9 fs were carried out in aerosol clouds of 10^{14} particles per cc and for fused silica, and cells containing water. Results indicated that there is a trend for greater transmission as the pulse length is shortened. The results indicated that the phenomena is just on the turn on of the electron not being able to follow the applied electric fields.

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FEMTOSECOND LASER PULSES THROUGH
AEROSOLS

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Introduction

Research was conducted to investigate the transmission of laser pulses through aerosol sprays to simulate propagation through clouds. Pulsewidths varied from continuous wave (cw) to as short as 9 femtoseconds (fs).

Measurements of the transmission through a confined aerosol cloud were performed using various femtosecond laser pulse lengths. In order to better understand the aerosol data, measurements of the transmission and reflection of femtosecond pulses of varying pulse duration incident on glass substrates was also carried out. These studies were performed to investigate the effect of pulse duration on the transmission and reflection in more detail on materials that were constant in time and would not have the variability that aerosols have. This report is organized into the following sections: theoretical considerations of femtosecond laser reflection and transmission, experimental setup and procedures, data results and interpretation, and conclusions.

Theoretical Considerations of Femtosecond Laser Reflection and Transmission

The propagation of light through a material induces a polarization of the material. For femtosecond pulses, electronic polarization becomes dominant because this is the only type of polarization that can respond this rapidly.

The response time of the linear polarization is assumed to be instantaneous in the Born-Oppenheimer approximation.¹ This approximation implies a lossless and dispersionless material.²

The response time of a nonresonant electronic nonlinearity was estimated to be 0.1 fs by estimating the time for an electron to orbit around an atom in the Bohr model.³ However, this value has not been measured to our knowledge and estimates of the electronic response time of 1 fs have been suggested.⁴

It is also known that the linear optical properties of materials tend towards unity at very high frequencies where the electrons can no longer respond to the field.⁵ A response time of 0.1-1.0 fs is a reasonable estimate of the linear electronic polarization response time in non-absorbing material.

The polarization which is induced in the sample determines the optical properties of the sample. For pulsed lasers, this transient change in polarization can be thought of as creating a time-dependent index of

refraction in nonresonant interactions as the polarization is induced by the laser pulse. For longer pulses, the transient change in polarization plays an insignificant role, but for a pulse of a few fs duration, the effect of the transient polarization needs to be examined. Fig. 1 shows a sketch of the transient nature of the index of refraction, $n(t)$, induced by the polarization and the associated reflectance and transmittance.

This transient change in the index of refraction means that the leading part of the laser pulse would have an enhanced transmission and reduced reflection loss at an interface since the electrons have not responded yet. Assuming a laser pulse width, τ , with constant intensity, an electronic response time, τ_e , of the material, and an instantaneous change in reflection and transmission occurring at a time, τ_e , after the arrival of the pulse, the transmission, T , as a function of τ_e and τ can be estimated as:

$$T = \tau_e/\tau + T_{ss}(1 - \tau_e/\tau) \text{ for } \tau_e < \tau \quad (1)$$

where T_{ss} is the steady state transmission predicted by Fresnel equations.

This is simply the sum of the transmission for each portion of the pulse (1 or T_{ss}), multiplied by the portion of the pulse energy in that portion of the pulse. Fig. 2 plots the transmission as given by Eq. 1 as a function of pulsewidth for 0.1 fs and 1.0 fs electronic response time and $T_{ss}=0.96$. This model predicts T to be from 0.04% to 0.4% higher than the cw case ($\tau_e = 0$)

for pulsewidths of 9 fs, the shortest pulse available in our lab. This work measures and compares the transmission of different pulse durations through aerosols and also solid substrates.

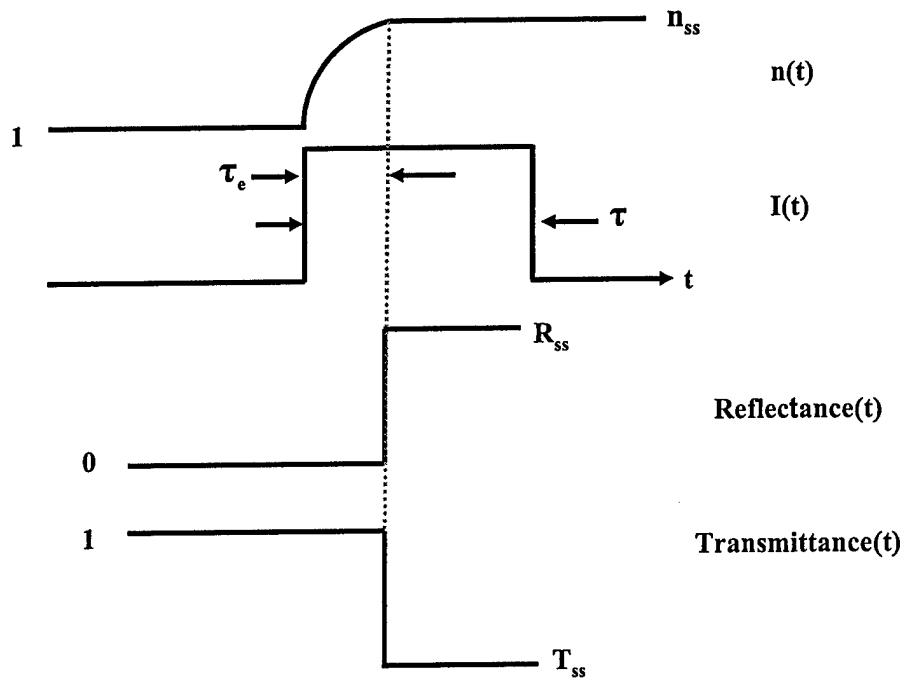


Figure 1. Diagram showing the time-dependent index of refraction of a material, $n(t)$, to a laser pulse with intensity $I(t)$ and pulsewidth τ . The index of refraction of the material changes from the initial value, assumed to be 1, to the steady state value, n_{ss} , on a time scale defined by the polarization response time, τ_e . The corresponding time-dependent reflectance and transmittance are also shown assuming that these both jump to a steady state value after a time delay of τ_e . T_{ss} and R_{ss} are the steady state reflectance and transmittance predicted by Fresnel's equations.

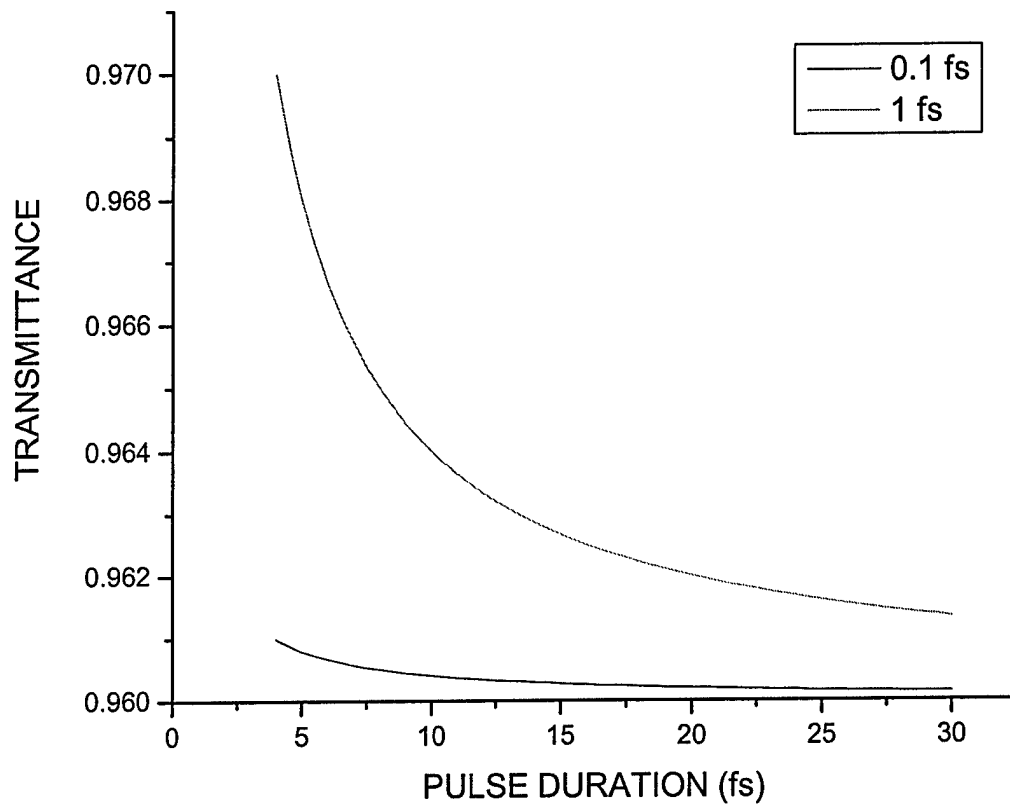


Figure 2. Calculated transmission of different laser pulsewidths through material with electronic response times of 0.1 and 1.0 fs.

¹ R. W. Hellwarth, *Prog. Quant. Electr.*, **5**, 1 (1977).

² R. W. Boyd, *Nonlinear Optics*, Academic Press, Inc., New York, 1992, p. 2.

³ R. W. Boyd, *Nonlinear Optics*, Academic Press, Inc., New York, 1992, p. 172.

⁴ P. Langot, S. Montant and E. Freysz, *Opt. Comm.*, **176**, 459 (2000).

⁵ M. Fox, *Optical Properties of Solids*, Oxford University Press, New York, 2001, p. 35.

Experimental Setup and Procedures

A schematic of the experimental setup is shown in Fig. 3 while a photo of the experimental apparatus is shown in Fig. 4.

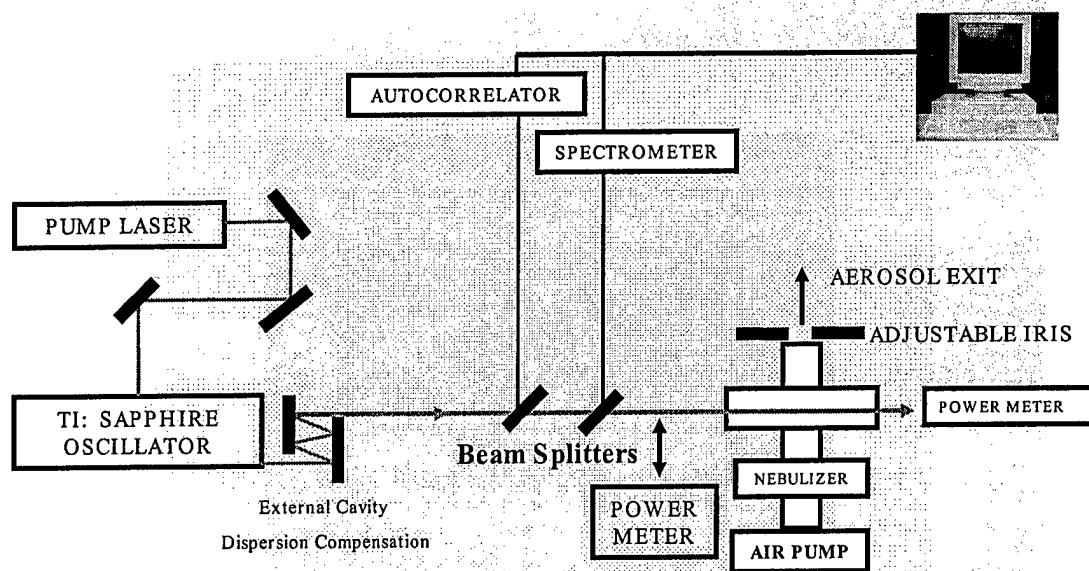


Figure 3. Aerosol transmission measurement setup.

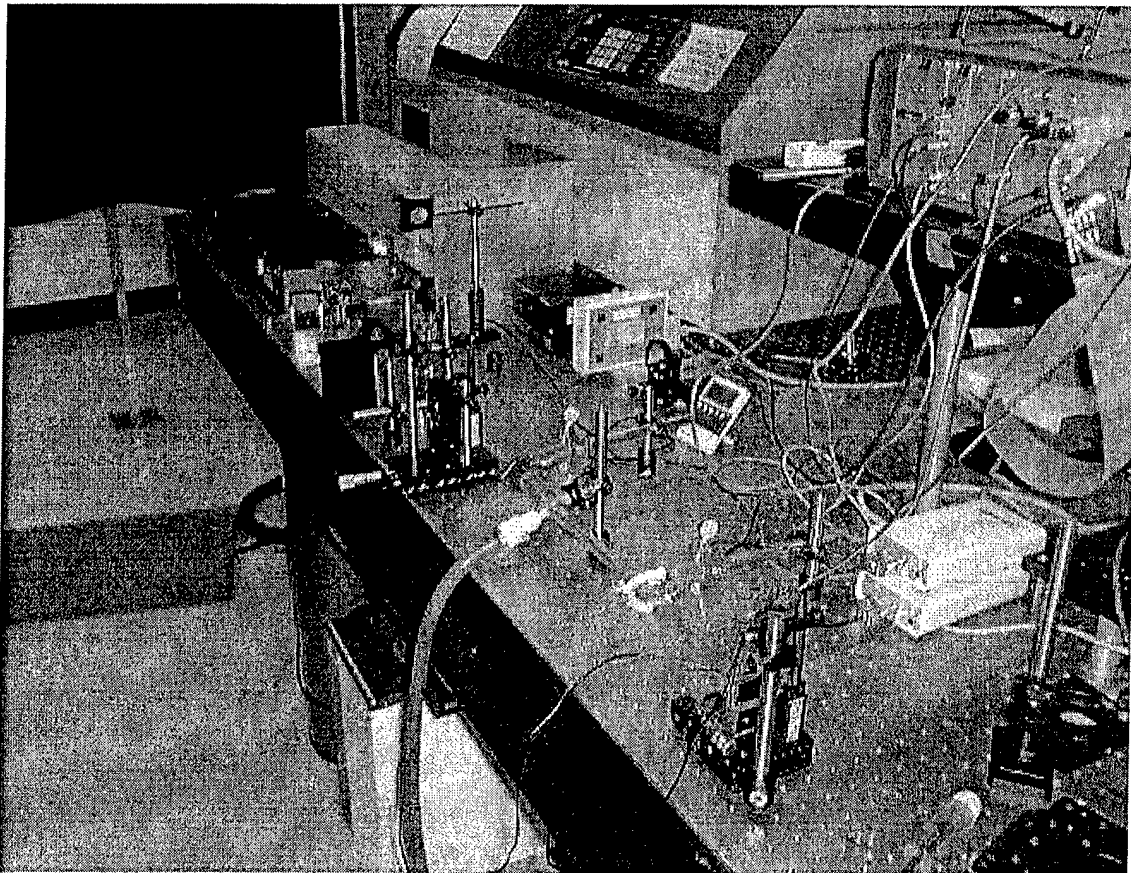


Figure 4. Photo of experimental setup.

The aerosol spray was generated by a Schuco Model 2000 air pump connected to a standard bronchial nebulizer filled with tap water. The aerosol spray was directed to a tee fixture with an adjustable iris opening which could be varied to get different transmission of laser light through the aerosol. The laser propagated in a direction perpendicular to the aerosol particle flow. Glass tubes extended from the tee along the direction of laser propagation to reduce turbulence in the aerosol. The length of the tubes was 15 cm on the laser entrance side of the tee and 30 cm on the exit side. An Aerometrics Phase Doppler Particle Analyzer (PDPA) was used to measure

particle size distribution at the exit of the tee as required. The typical particle size distribution and related statistics are shown in Fig. 5.

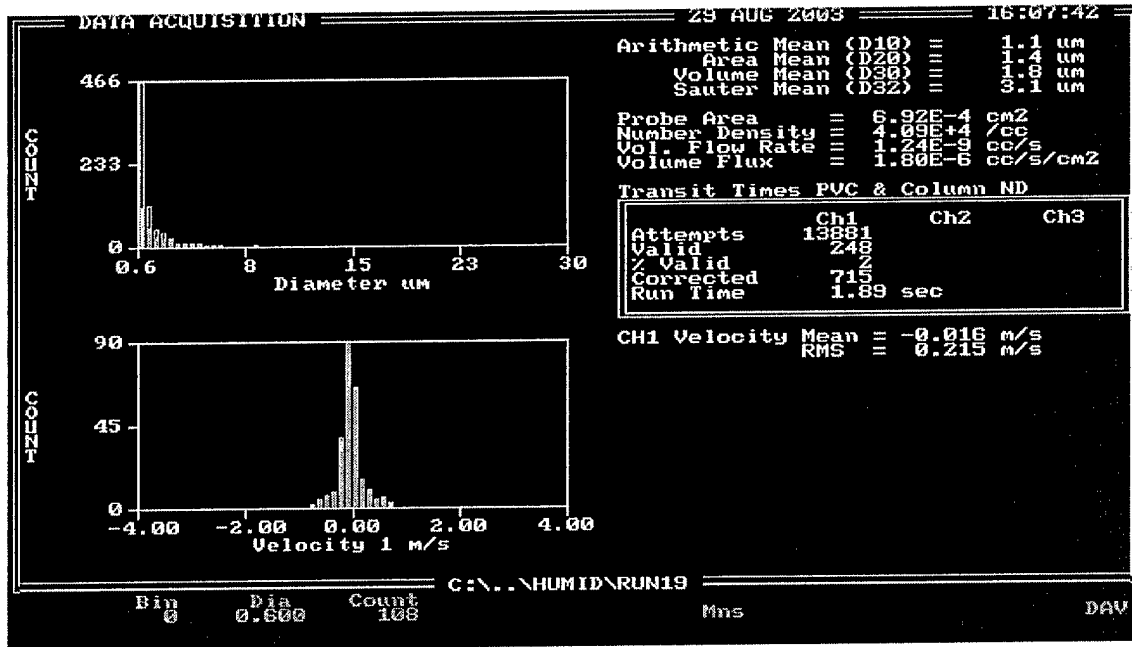


Figure 5. Typical nebulizer particle size distribution. Number density varied from $3\text{-}5 \times 10^4/\text{cc}$ in 6 data runs over 20 minutes.

The femtosecond laser pulses were produced by a Femtosource Compact laser manufactured by Femtolasers and pumped by a Millenia X visible laser made by Spectra-Physics. External Cavity Dispersion Compensation mirrors are used to compensate for dispersion in the output optics of the laser as well as the substrate of the 2% beamsplitter. This beamsplitter directs the beam to the autocorrelator (Femtometer manufactured by Femtolasers) used for pulsewidth measurement. Inside the autocorrelator, pulses have one pass through the same material that the beam splitter is

made of, so the pulse duration measured is the same as the pulse which exits the 2% beamsplitter. A 0.7% beamsplitter sends light to a spectrometer (Avantes S2000) to measure the spectrum of the laser pulse. Both the autocorrelation and spectrum are displayed on a computer where the full width half maximum (FWHM) pulsewidth is calculated assuming a sech^2 laser pulse shape. Fig. 6 shows the typical autocorrelation trace and spectrum from the laser.

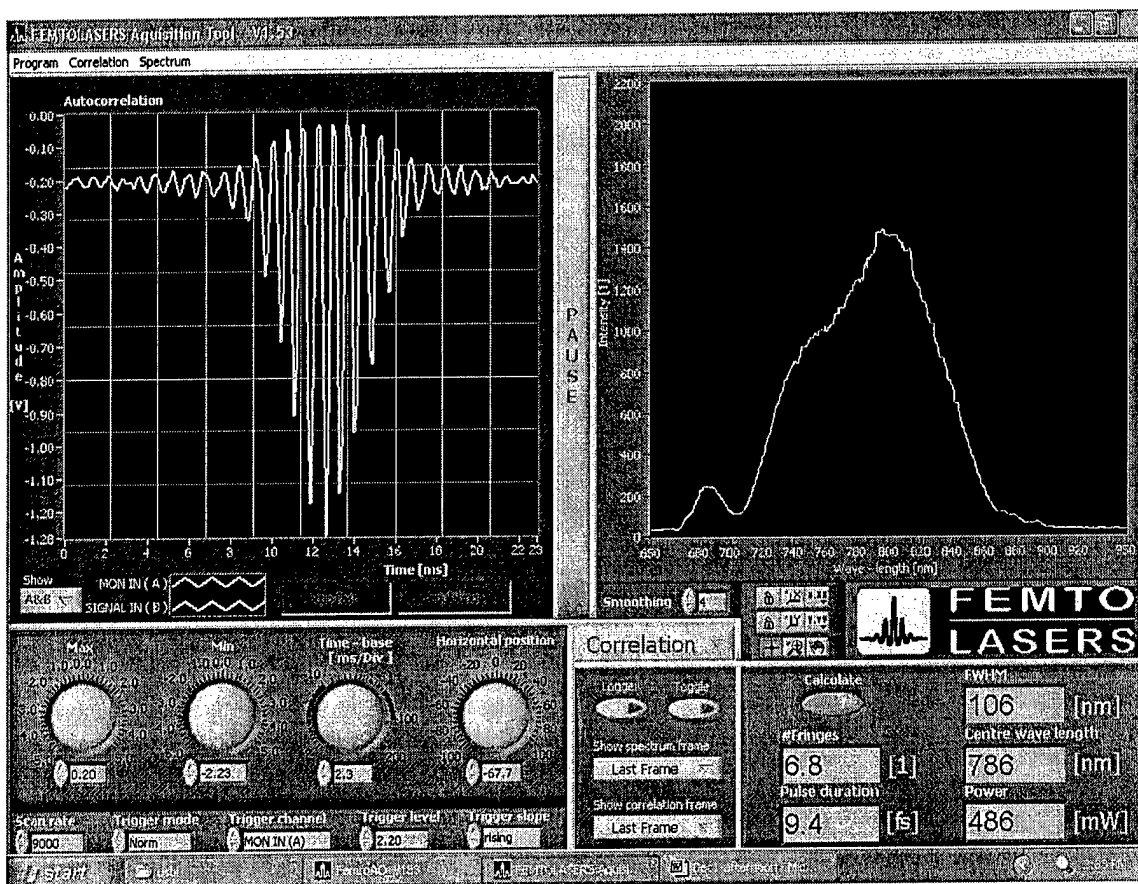


Figure 6 Typical autocorrelation trace and spectrum from the laser.

The incident and transmitted power through the aerosol cloud is measured using a thermal detector (Ophir, 30A). The output of the detector head is connected to a USB interface (Ophir) to interface with a computer. Power data can be collected at 15 Hz with this system.

In order to investigate the pulsewidth dependence of the transmission and reflection with more accuracy, measurements were made on fused silica samples. These fixed measurements allowed higher accuracy measurements to be made since the inherent fluctuations in an aerosol are eliminated and a reflected and transmitted power data was collected simultaneously which reduced errors due to laser power drift. The experimental setup used in performing these measurements, is shown in Fig. 7. Photodiodes with attenuating filters (PD-300-3W, PD300-SH from Ophir) were used to measure the transmitted and reflected power in this case. A USB interface with the detectors allowed data to be collected on a computer.

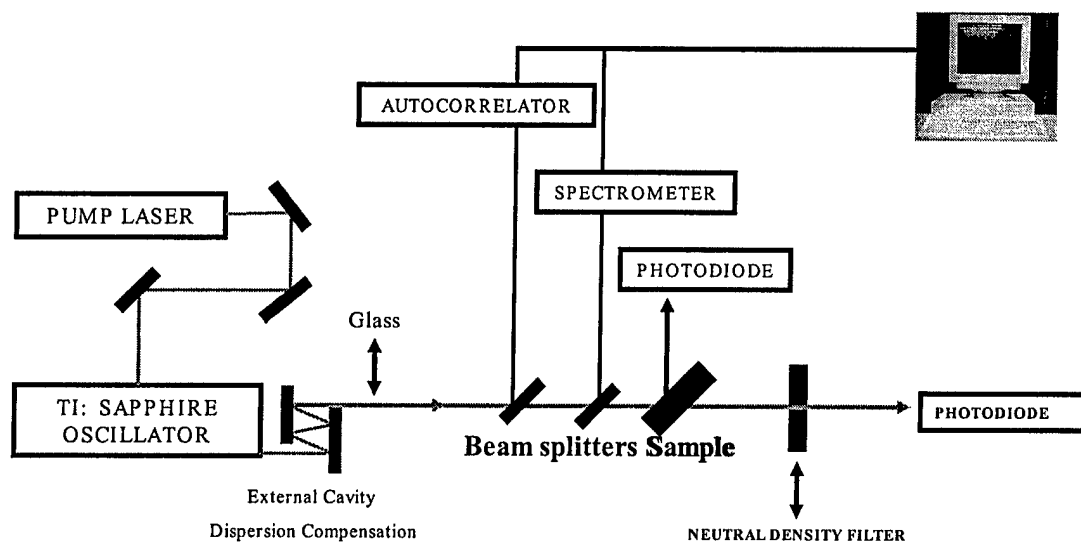


Figure 7. Experimental setup for reflection and transmission measurements.

Data Results and Interpretation

Initial experiments compared transmitted laser power through an aerosol cloud for 12 fs pulses and continuous wave (cw) cases. The 12 fs pulsewidth results when using both beamsplitters in the path. This arrangement makes it possible to have both the spectrum and the autocorrelation data information about the pulse, and can be shortened to 9 fs by removing the second beam splitter. The Femtolaser laser was switched between mode-locking and cw for these comparisons with a center wavelength near 800 nm in each case. The beam diameter changes between mode-locked and cw operation of the laser and, therefore, a 1 mm diameter iris was inserted in the beam path before transmission through the aerosol to minimize any non-uniform spatial sensitivity on the thermal detector head. The statistics of the measured transmission difference between modelocked pulses and cw is shown in Fig. 8. The percent difference between modelocked and cw transmission was calculated by subtracting the cw transmission percentage from the modelocked transmission percentage. This data represents data taken over 8 trials. Each trial had 20-40 data runs, each consisting of measuring transmission through the aerosol for both modelocked and cw cases. The average of 20-30 seconds of data for each data run was recorded and the percent transmission calculated for each case.

Nominal transmission values from 60-85% were created by changing the iris diameter at the aerosol exit. No correlation on the aerosol density was noted between mode-locked and cw transmission.

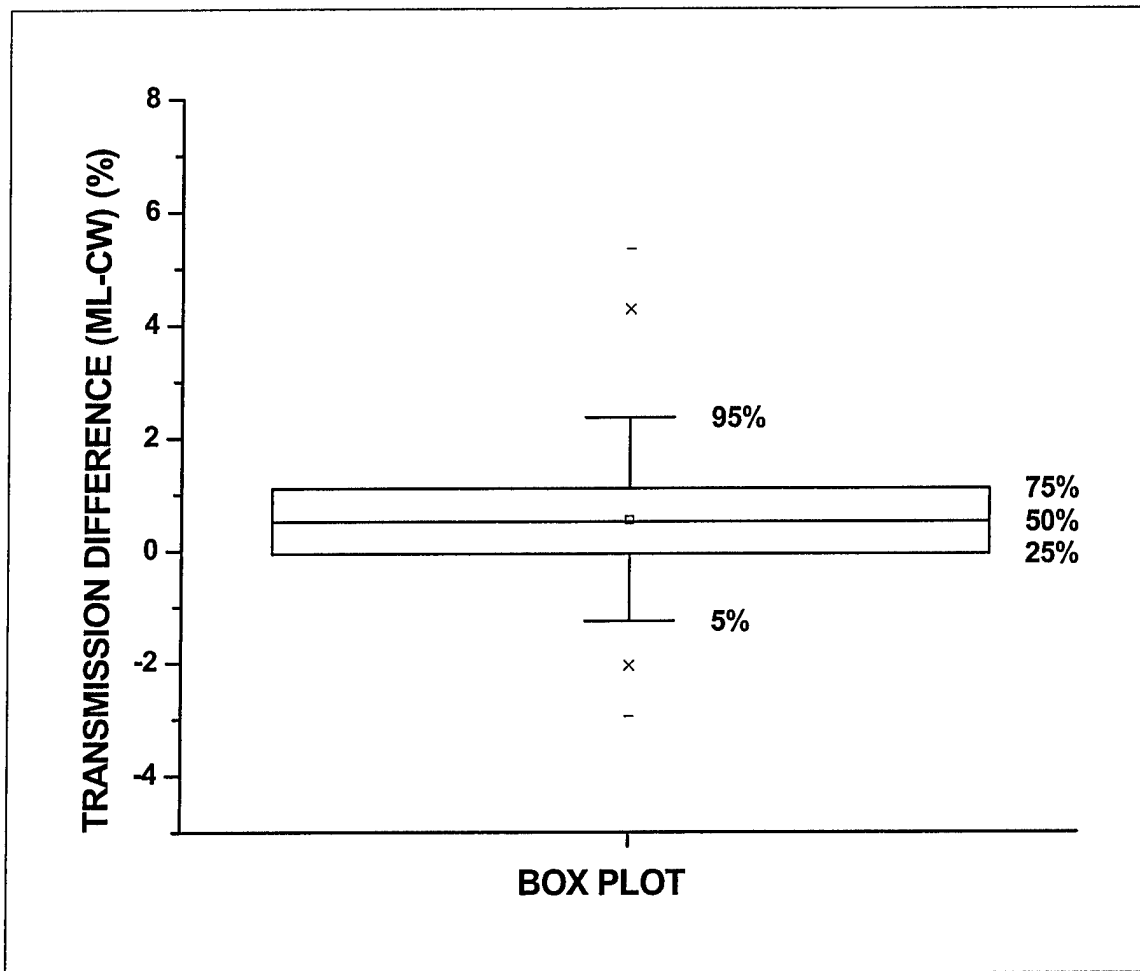


Figure 8. Box plot of statistics of the transmission difference between 12 fs mode-locked (ML) pulses and cw laser propagating through aerosol. 12 fs pulses had a mean value of transmission which was 0.5% larger than cw. Maximum and minimum are represented by dash and 99% and 1% ranges are represented by x's.

The spread in the data is caused by the variations in density in the aerosol and the inherent variations in the laser output power of $\sim 0.3\%$. The accuracy of the data is also limited by the linearity of the thermal head

detector which is specified at $\pm 1\%$ with power. Spatial variations were minimized by moving the detectors to the same position on a translation stage.

Since the increase in transmission caused by the finite polarization response time is believed to be general and should be true for other materials, other samples were also investigated. To improve measurement accuracy, photodiodes which have excellent linearity vs. power, were used instead of the thermal detector heads. Simultaneous collection of transmitted and reflected power from the sample also helped to reduce errors due to drift of the laser power. The iris is also eliminated in this setup because the laser beam size change when switching between modelocked and cw is no longer an issue when only modelocked pulses are used. Elimination of the iris removes errors which could be caused by differences in the diffraction of the modelocked and cw pulses and the associated spatial pattern on the detector. The ratio of the transmitted to reflected power was then compared for different pulse durations on different samples and compared to theoretical predictions.

Solid samples consisting of different glasses and liquid water contained in thin walled containment cells were used to eliminate the variability thought

to be caused by the aerosol cloud. The transmission and reflection of fused silica, an empty fused silica cuvette, and water in the fused silica cuvette were measured with different laser pulse widths to gain a better general understanding of the pulsewidth dependence of the transmission in other materials. The pulsewidth was stretched by inserting fused silica windows varying from 1 to 6.3 mm thickness in the beam path as shown in Fig. 7. The wedge angle between the two faces of the window used to stretch the pulse was less than 10 seconds for minimal deviation as the beam passes through. The sample was a 2 mm thick fused silica window. The angle of incidence on the sample was nominally 10 degrees and power reflected from both surfaces of the sample and the power transmitted through the sample was detected simultaneously using photodiodes. The second beamsplitter in the setup shown in Fig. 7 was removed when collecting data to eliminate pulse stretching in this optical element and obtain the shortest pulses possible.

Figs. 9-11 show the change in the ratio of the transmitted power over the reflected power (P_t/P_r) normalized to the ratio at 10 fs. A downward trend in P_t/P_r was observed for 75% of the data while the other 25% showed no clear trend. The two sets of data points shown in Figs. 9-11 represent the maximum and minimum drop observed in P_t/P_r over the 10 to 20 fs range for

the data with a downward trend in P_i/P_r , while the line is the calculated value using Eq. 1.

In Fig. 9, calculated values of T/R using Eq. 1 for a response time of 0.01 fs and steady state transmission of 0.96 are also shown to compare to the measured data. The pulsewidth dependent reflectance, R , is calculated as $1-T$ from Eq. 1. This value is smaller than the 0.1-1 fs response time estimated by theory. The most likely reason for this discrepancy is that the model developed overestimates the changes in transmittance and reflectance for a given polarization response time because of the assumptions of an instantaneous change in the transmittance and reflectance.

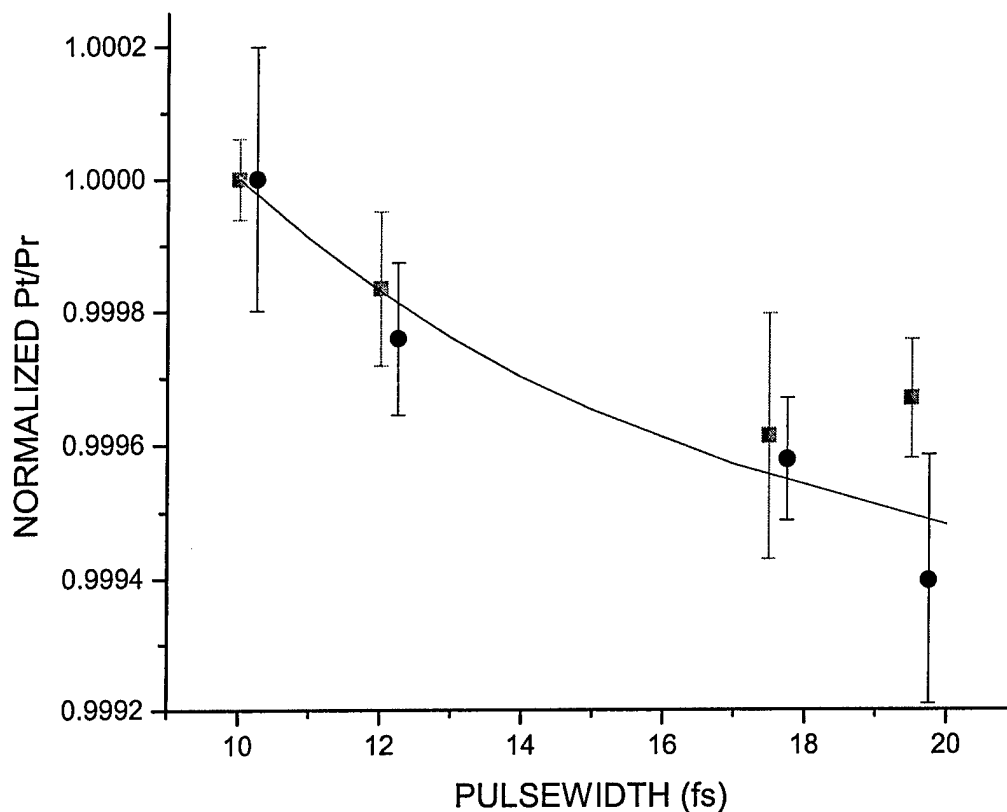


Figure 9. Measured and calculated transmitted power/reflected power from a fused silica sample. Values were normalized to the 10 fs pulse. The two data sets (taken on different days) show the largest and smallest drops in P_t/P_r for the cases where a clear downward trend in the data was observed.

Figs. 10 and 11 show the normalized transmittance/reflectance ratios of an empty fused silica cell and the same cell filled with water. The theoretical curves shown for comparison in Figs. 8 and 9 are for 0.04 and 0.03 fs response times respectively. More variation in the data is observed in these cases.

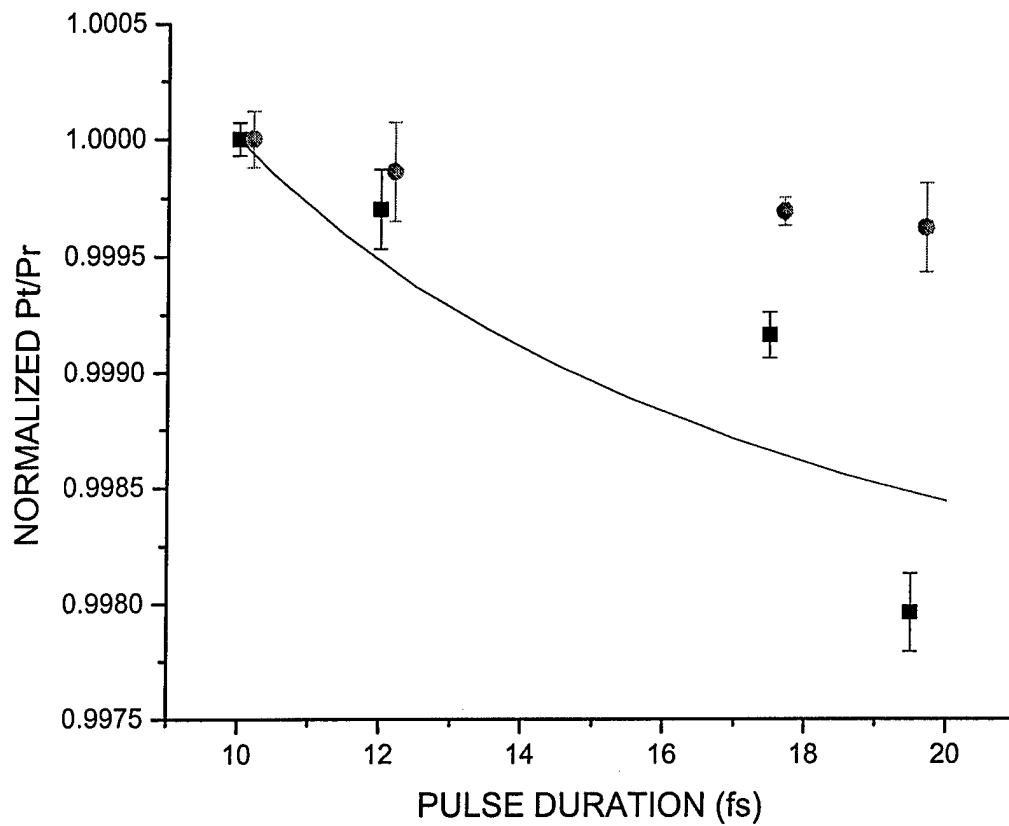


Figure 10. Measured and calculated transmitted power/reflected power from a fused silica sample cell. The two data sets (taken on different days) show the largest and smallest drops in P_t/P_r for the cases where a clear downward trend in the data was observed.

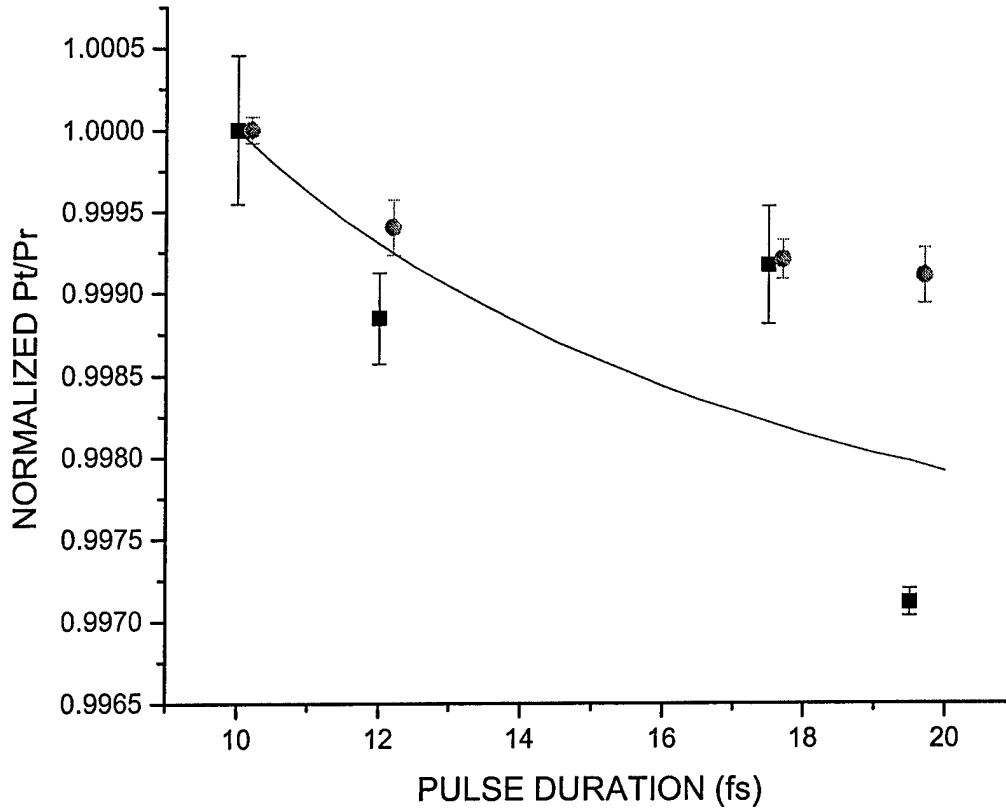


Figure 11. Measured and calculated transmitted power/reflected power from a fused silica sample cell filled with water. The two data sets (taken on different days) show the largest and smallest drops in P_t/P_r for the cases where a clear downward trend in the data was observed.

The effect of multiple interfaces in the experiment, rather than the single interface assumed in Eq. 1 complicates the analysis. Pulse stretching due to dispersion in the different materials also needs to be considered. Fig. 12 shows these effects for several reflections from a sample. More interfaces actually occur in the experimental setup including neutral density filters and detector windows. Modeling of multiple interface effects and pulse stretching caused by dispersion was not attempted in this work.

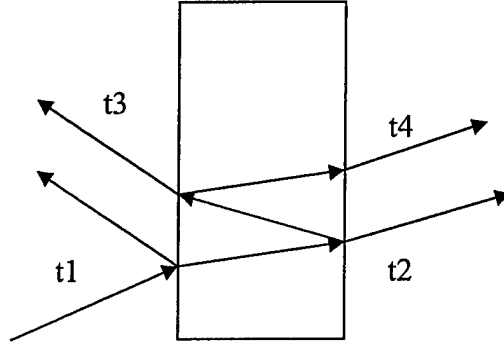


Figure 12. Diagram showing multiple reflections and pulse stretching due to dispersion in a material. Incident pulse with pulsewidth t_1 passes through the material and emerges with a pulsewidth of t_2 . Secondary reflections exit the material with pulsewidths t_3 and t_4 .

Nonlinear Effects

Nonlinear effects are considered to understand how they may contribute to the measured transmission and reflection. The intensity, I , of the pulses can be expressed as:

$$I = \frac{E}{\pi r_0^2 \tau} \quad (2)$$

where E is the pulse energy, r_0 is the beam radius and τ is the pulsewidth.

Calculating the intensity for the laser for pulse energy of 5 nJ, 1 mm beam diameter and 9 fs pulsewidth gives $I \sim 10^8 \text{ W/cm}^2$. Nonlinear effects have been measured for pump powers of 10^8 W/cm^2 for 100 fs pulses.⁶

The nonlinear index of refraction, n_2 , can be expressed as:

$$n_2 = \frac{0.0395 \chi^{(3)}}{n_0^2} \quad (3)$$

where $\chi^{(3)}$ is the third-order nonlinear susceptibility, and n_0 is the linear index of refraction. n_2 of fused silica (measured at 800 nm) is $6 \times 10^{-16} \text{ cm}^2/\text{W}$, giving a value of $n_2 I \sim 6 \times 10^{-8}$ for $I = 10^8 \text{ W/cm}^2$.⁷ The nonlinear index of refraction for water has a similar value.⁸ Comparing the normal incidence Fresnel reflectivity of fused silica, an increase by $\sim 10^{-8}$ in the reflectivity is calculated for an increase in the refractive index of 10^{-7} caused by nonlinear effects.

Average powers used in the experiment were approximately constant, although the effect of stretching the pulse will decrease the peak intensity of the pulse. Note that an increase in reflectivity due to nonlinear index changes is predicted for higher intensities (or shorter pulses given a fixed average power), opposite of what was observed.

Saturable absorption in the absorbing neutral density filters integrated into the photodiodes is another nonlinear effect which could appear in the data.

Obtaining data at lower powers and using photodiodes without neutral density filters will eliminate these possible nonlinear effects.

⁶ J.-K. Wang, T.-L. Chiu, C. H. Chi, and C.-K. Sun, *J. Opt. Soc. B*, **16**, 651 (1999).

⁷ J.-K. Wang, T.-L. Chiu, C. H. Chi, and C.-K. Sun, *J. Opt. Soc. B*, **16**, 651 (1999).

⁸ B. A. Rockwell, W. P. Roach, M. E. Rogers, M.W. Mayo and C. A. Toth, *Opt. Lett.*, **18**, 1792 (1993).

Conclusions

Measurements of the transmission of 12 fs modelocked pulses and cw laser light through a confined aerosol showed a transmission of 0.5% more for the 12 fs pulses. Aerosol densities were adjusted using an iris to give transmission values of 60-85%.

Additional experiments designed to improve the accuracy of the transmission and reflection measurements were performed on fused silica samples and water in a sample cell. Measurements of the transmitted and reflected power were performed and the ratio P_t/P_r was measured as pulsewidth was varied from 10-20 fs for these samples. A downward trend in P_t/P_r as pulsewidth increased occurred for 75% of the data while no clear trend was seen in the other 25% of the data. The downward trend is in agreement with a transmission model which is dependent on pulsewidth and electronic polarization response time. Values of the response time which gave reasonable agreement with the data varied from 0.01 to 0.04 fs, smaller than previous estimates of 0.1-1 fs. More complex models which incorporate finite polarization response times and non-instantaneous changes in transmission and reflection could be developed to improve agreement. Multiple interfaces and pulse stretching due to dispersion are other effects which should be included in more sophisticated models.

The experiments carried out starting at about 10 fs showed that there is a small trend for ultra short pulsed to pass through a material without interaction. Since the phenomena is just at the turn on there is a need to perform further research at short pulse lengths. Thus, further research is recommended where pulses of 4 fs or shorter are used to conduct similar research to better understand the phenonomena.